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THE CRITICAL TEMPERATURE-CONCENTRATION PHASE DIAGRAMS IN SOME KONDO AND SPIN GLASS ALLOYS

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ABSTRACT

Competitions between Kondo, spin glass, Ferro, antiferromagnetic, and paramagnetic phases in magnetic alloys are very complicated problem. Experimentally, in most magnetic systems, thermal critical points like Kondo temperature, freezing temperature, Curie and Neel temperature, maximum and minimum temperatures could be considered as characteristic points for each phase of these types. Theoretically, in magnetic alloys, behavior of exchange integral constantJ_{xx} (xx = S-d, d-d, and F-F represent types of spin interactions) could be directly understood through linked it with these critical points. Each magnetic alloy with some magnetic impurity concentration has more than critical temperature, but each magnetic critical concentration has its own phase boundary between phases, and converts magnetic regime to another regime. A conflict between theoretical and experimental studies has been found, because spin glass and Kondo theory has not yet exactly completed.

KEYWORDS: Kondo, Freezing, Curie, Neel, Maximum and Minimum Temperatures

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INTRODUCTION

The aim of this paper is to find out the behavior of the critical temperatures as a function of concentrations in a special kind of magnetic alloys, and compare these cortical temperatures with magnetic exchange integral through identical relations. It will be, magnetically, compared between two types of alloys (Au-Fe and Cu-Mn), each of them consists of noble metals as a lattice and a few percentages of 3d transition metals as impurities on some sites of the noble metal lattice, where sites and bonds distribution of 3d metals on the lattice is randomness, that is a basic condition of these alloys.

Thermal transition investigations carried out on Au-Fe, Cu-Mn alloys, with a concentration range from 10 ppm till more than 17% per magnetic impurity, and thermal spectrum from 0.1 to 300 K.

The first alloy (Au-Fe) begins from Kondo alloys to spin glass alloys and end with ferromagnetic regime, but the second alloy (Cu-Mn) ends with an antiferromagnetic case with increasing magnetic impurity. Figure (1) illustrates how these magnetic phases distribute according to magnetic impurity concentrations [1-3].

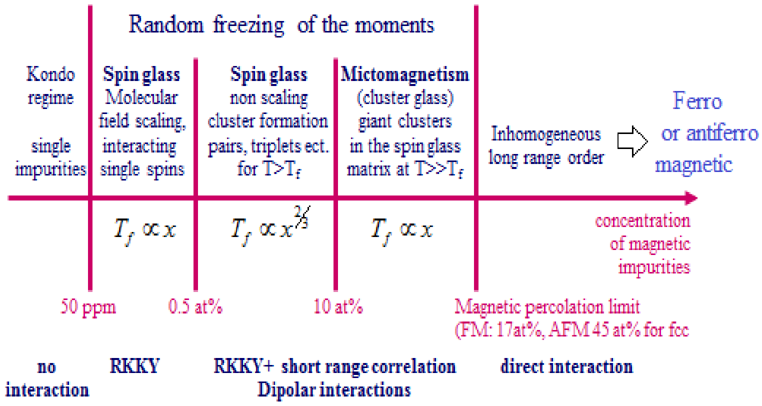

Figure 1: Concentration Regines for Transition Metals (After Sollerhaus 2002: Stephan Mair)

The problem of Kondo regime was discovered by Kondo in 1964[4], and characterized by s-d exchange interactions[5], whereas spin glass is a new magnetic system discovered in 1972 [6], and characterized by indirect exchange interactions of localized spins via conduction electrons or The Ruderman – Kittel – Kasuya - Yosida (RKKY) interactions[7 -10], RKKY interactions is one of the important mechanisms explaining the origin of magnetism in some solids and distinguish between spin glasses and another magnetic systems.

Ferro and antiferromagnetic regimes characterized by direct interactions between localized spins and by Heisenberg and Isingmodels [11 -15].

In this paper, the competitions between these regimes will be examined, and the results will depend on many thermal transition factors which will help to give a better picture about boundaries of each regime.

QUANTUM THEORETICAL BACKGROUND

Theoretically, concentration of magnetic impurity in magnetic alloys is responsible about which theoretical model will be used, so that, figure (1) shows that many theoretical models have to be used, where may be divided as follows:

- Kondo model depends on s – d exchange interaction between the spins of the conduction electrons S_e and the single localized magnetic moment S_d of the transition element, which can be described by a Hamiltonian[5]:

$$\mathrm{H} = J_{sd} S_e . S_d \tag{1}$$

Where ($\mathbf{J_{sd}}$) is the exchange coupling parameter, and has always negative values in Kondo regime.

- Edwards-Anderson (EA) model for Spin glass [16] was the first theoretical model, where followed by the model of SK(Sherrington & Kirkpatrick)[17,18], Each of them described the state of spin glass according to the following Hamiltonian:

$$\mathrm{H} = \sum J_{ij} S_i . S_j \tag{2}$$

Where $\mathbf{S_i}$ is the spin in position i, $\mathbf{S_j}$ is the spin in position j, $\mathbf{J_{ij}}$ is an indirect exchange constant between localized magnetic moments via the host conduction electrons and its RKKY energy has the following form:

$$J(R_{ij}) = \frac{9\pi n^2}{E_F} J_{sd}^2 \frac{x\cos x - \sin x}{x^4} \tag{3}$$

Where $\left(x = 2k_F R_{ij}\right)$, $\{J_{ij} = J\left(R_i R_j\right) = J(R_{ij})\}$, EA, SK and others had described the system through an order parameter carries qualities of remembrance (memory) at its critical (freezing) temperature [19, 20]

- Heisenberg model[11] for Ferro and antiferromagnetic depend on direct interactions between adjacent spins according to the following Hamiltonian:

$$\mathrm{H} = -\sum_{i,j} J_{ij} S_i . S_j \tag{4}$$

J_{ij} is exchange integral (its unit is energy unit) for entire lattice $J_{ij}=J_e$=constant, then the summation over all the atoms in crystal give:

$$\mathrm{H} = -J_e \sum_{i,j} S_i . S_j \tag{5}$$

WhereJ_e is the coupling constant or exchange integral,J_e is postive for ferromagnetic metals,whreas J_e is negetive for antiferromagnitic materials.

Exchange interaction constant (j_{ij}) is considered a cornerstone between theoretical and experimental results in all regimes.

When spins are only directed with z direction, then interactions at Cartesian axes (x, y) may by neglected, and then the Hamiltonian is just:

$$\mathrm{H} = -J_e \sum_{i,j} S_{zi} . S_{zj} \tag{6}$$

This is called Ising model [12].

- RKKY Hyperfine interactions [21]where nuclear spins S_n begin to interaction with 3d spins S_d at very low temperature, if we consider the hyperfine spin is ($F_i = S_d + S_n$) for magnetic impurity at the site i, we can obtain the Hamiltonian of the hyperfine RKKY interactions in spin glass regime:

$$H_{hf}^{RKKY} = -\frac{1}{2}\sum_{ij} J_{ij} F_i F_j \tag{7}$$

Hyperfine interactions in Ferro and antiferromagnetic will become directed interactions and the Hamiltonian then:

$$H_{hf} = -\frac{1}{2}\sum_{ij} J_{ij} F_i F_j \tag{8}$$

According to these theoretical models, Kondo effect was extensively treated [5, 22] and SK model predicted that, phase transitions have a sharp maximum of magnetic susceptibility and specific heat capacity at the freezing temperature. This model could not give a good interpretation of these anomalous phenomena. Figure (2) shows the theoretical outline of the magnetic phase diagram according to SK theory, where many models conflicted with SK model [23- 25].

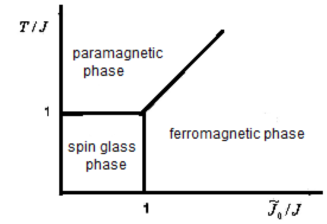

Fig.(2): Theoretical magnitic phase diagram of spin glass regime
(Chiu Fan Lee and Neil F. Johnson , ar Xiv: quant-ph/0412069 v1 8 Dec 2004)

The quantum theory of spin glass does not provide a successful solution, and the so-called phenomenological model of spin glass has emerged [26 -31] which examines the phenomenon through spin relaxation times and their relationship with the critical temperatures.

The relationship between theoretical and experimental studies depend on the relation between exchange coupling constant and some critical temperatures where magnetic phase changes from one phase to another.

The trend of this paper will consider every exchange coupling constant J_{xx} is identical with one of critical temperatures $T_{critical}$ as a function of critical concentration C_c, and take the following form:

$$J_{xx} \equiv kT_{critical} = f\ (c_c) \tag{9}$$

EXPERIMENTAL DATABASE

Critical temperatures like, Kondo temperature T_k (the temperature above which Kondo effect loses its character), freezing temperature T_f (the temperature above which a spin glass becomes super paramagnetic or ferromagnetic), Curieor Neel temperature T_c (the temperature above which a ferromagnetic or antiferromagnetic substance loses its magnetism and becomes paramagnetic.), maximum temperature T_{max} means that phonons spread in samples, and minimum temperature T_{min} means that phonons or magnons spread in samples. All above temperatures were investigated, and collected from many sources[32-41],Because the urgent need to many sources, like resistivity, specific heat, susceptibility, Mössbauer effect, neutron scattering, magnetization and hysteresis cycles, NMR, and SR, to give a large number of Critical points. Dynamic studies showed that there was no constant phase transition in the spin glass system. Figure (3) shows some windows for relaxation times companioned with spin freezing temperatures [42, 43],where external magnetic fields, may give very long relaxation times (field cold & zero-field cold) [44].

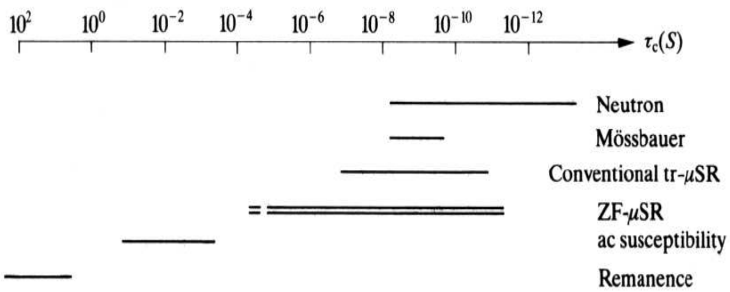

Figure 3: Windows to See Spin Glass Dynamical of Different Relaxation Times (After Uemera, Et.Al, 1985)

RESULTS AND DISCUSSIONS

Two kinds of critical points may be distinguished in magnetic systems; the first is the critical concentration where the behavior of regime transforms to another regime, and the second is the critical temperature where there are also two kinds, the first one is the critical temperature belong to critical concentration, and the second is the critical temperature belong to any magnetic alloy itself which has some magnetic impurity concentration in the same regime. In other word every magnetic alloy has its critical temperatures in the same regime.

According to figure (1), critical points for all regimes may be classified as follows:

- **Kondo Temperatures** correlated with Kondo alloys and their magnetic impurity concentrations begin from a few ppm (part per million) of magnetic impurity till a critical concentration where another regime will prevail.

Kondo regime meeting with spin glass regime at a critical point $T_f^c = T_k^c$, and J_{sd} might be written as an identical or equivalence form:

$$J_{sd} \equiv kT_k = f\ (c) \tag{10}$$

Where f (C) is a function of concentration, Figure (4) Shows the Behavior of Kondo Temperatures with Concentrationfor Cu-Mn Alloys

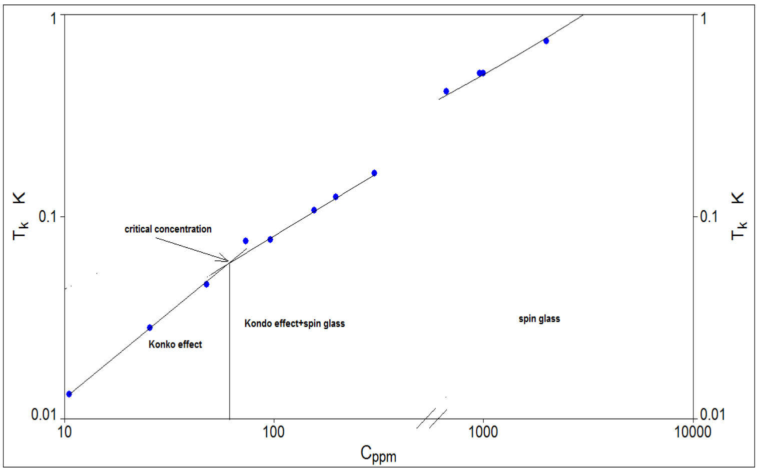

Figure 4: Critical Kondo Temperatures as a Function of Ppm Concentrations in Cu-Mn Alloys

Theoretically, (T_k) has the following phenomenological relationship:

$$Ln\,T_K = Ln\,T_F - \frac{1}{n(E_F)\left|J_{sd}\right|} \tag{11}$$

Where, initially, T_F =8.12 10^4 k, and n (E_F) =0.294 ev^{-1}, J_{sd} was calculated [5] as a function of concentration for Cu-Mn Kondo alloys and equals to:

$$J_{sd} = -0.014898811 Ln\,C - 0.17956871 = f(c)\ ev \tag{12}$$

Figure (5) shows the relationship between Kondo temperatures andex change constant and critical point between Kondo and spin glass regime for Cu-Mn alloys.

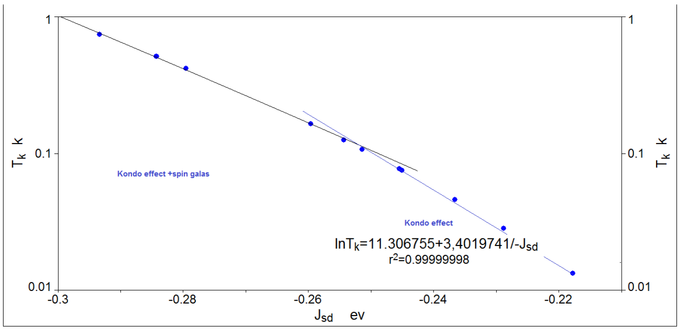

Figure 5: Critical Kondo Temperatures as a Function of Exchange Coupling in Cu-Mn Alloys

- **Freezing Temperatures** belong to spin glass alloys, and most values of spin freezing temperatures came from magnetic susceptibility, where at critical freezing temperatures, susceptibility denotes to a sharp cusp maximum in its value[45], and a new phase transition happening at which spin glass occurred. There are three kinds of spin glass:
- Ideal spin glass begins from nearly tens of ppm till 1% of magnetic impurity concentration where ideal RKKY interactions between single spins will prevail and critical temperature which is freezing temperature will take the following form :

$$J_{RKKY} \equiv kT_f = f\,(c_c) \tag{13}$$

According to figure (3) T_f does not represent a critical magnetic phase because it has many windows and many different relaxation times.

And from phenomenological models, the ideal spin glass phenomenon is described through spin relaxation times and their relationship with freezing temperature according to Arrhenius law:

$$\tau = \tau_0 \exp\left(\frac{E_a}{K_B T_f}\right) \tag{14}$$

- The real spin glass begins from 1% to about 10% of magnetic impurity concentration, RKKY interactions will be between clusters, and freezing temperature will stop at T_0:

$$J_{RKKY} \equiv k\,(T_f - T_0) = f\,(c) \tag{15}$$

T_0 is the temperature at which RKKY interactions are almost complete, and real spin glass follows phenomenological Vogel-Fulcher law:

$$\tau = \tau_0 \exp\left(\frac{E_a}{K_B\,(T_f - T_o)}\right) \tag{16}$$

Where, τ0=10-11 Sec, and Ea is activation energy, and this model has given results exceeded the theoretical

description [46, 47], and proved that the spin glass system does not pass through a phase transformation but is in the similar situation of viscosity in ordinary glass.

- Mictomagnetic (clusters spin glass) a new regime called re-entry (reentrant) in spin glass regime [48], or the mixed system (micto-magnetic) of spin glass with ferromagnetic or antiferromagnetic (SG+FM or AFM) appears when the concentration ratio exceeds the real spin glass system. Concentrations usually begin from about 10% to 17% of magnetic impurity concentration where some magnetic clusters interactions with each other by RKKY interactions, here there are intra interactions inside each cluster, and inter interactions between clusters across RKKY interactions. It's a very complicated situation because there are two critical temperatures, freezing temperature and Curie –Wiese temperature:

$$\begin{aligned} J_{RKKY} &\equiv kT_f = f(c) \\ J_e &\equiv kT_c = f(c) \end{aligned} \tag{17}$$

But Ferroor antiferromagnetic prevail where magnetic impurity concentration is more than 17% and a critical temperature will be only Curie –Wiese temperature:

$$J_e \equiv kT_c = f(C_c) \tag{18}$$

Figures (6, 7) show the behavior of freezing temperatures as a function of the concentration of magnetic impurities with a wide range of concentration, where Cu-Mn alloys tend to antiferromagnetic, but Au-Fe tend to ferromagnetic regime.

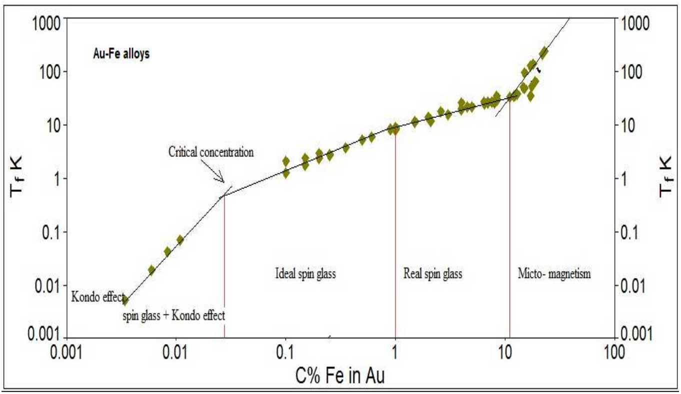

Figure 6: Critical Freezing Temperatures as a Function of Concentrations in Au-Fe Alloys

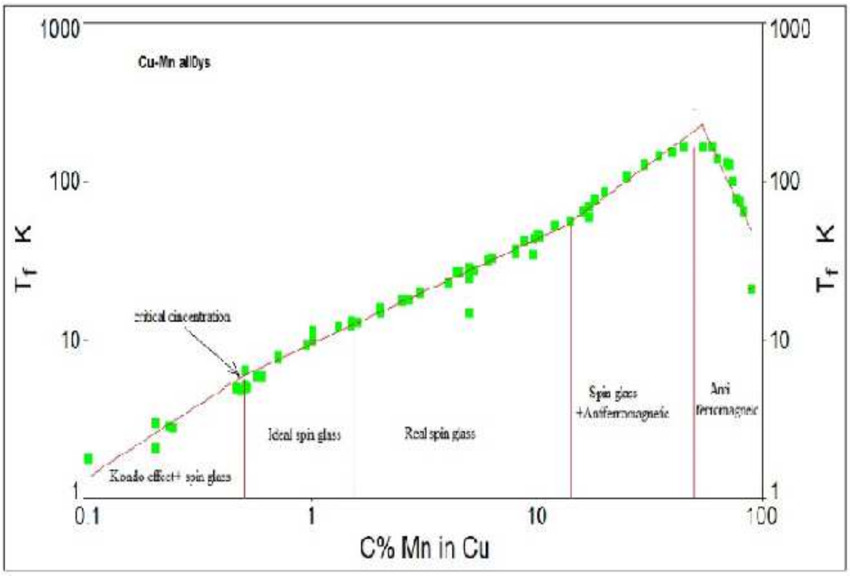

Figure 7: Critical Freezing Temperatures as a Function of Concentrations in Cu-Mn Alloys

- **Maximum Temperatures** come fromstatic studies, like electrical resistivity and the specific heat capacity, whichgive a broad maximum at T_{max} [49 - 51]where a broad maximum extended to long ranges of temperatures, showed no appearance of a phase transformation. Above these temperatures alloys subject to Curie's law and this system in paramagnetic phase (PM). Figures (8, 9) showed better fit between the temperature of the maximum resistivity and wide range of magnetic impurity concentrations.

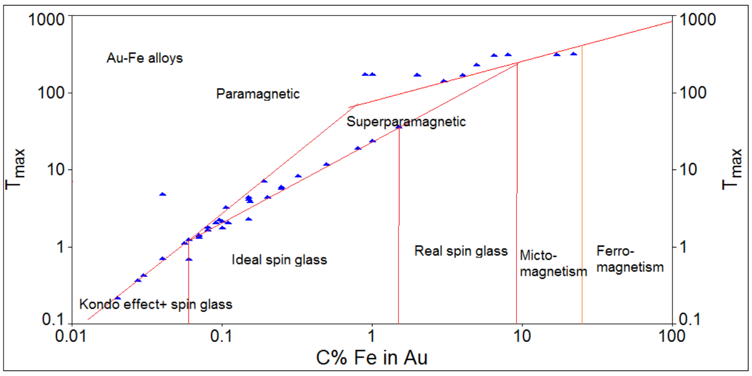

Figure 8: Critical Maximum Temperatures as a Function of Concentrations Au-Fe Alloys

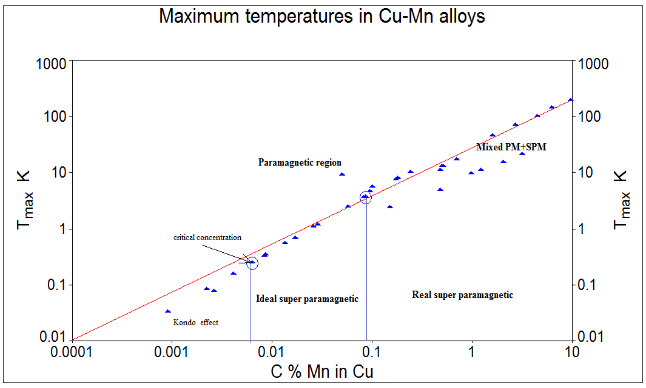

Figure 9: Critical Maximum Temperatures as a Function of Concentrations Cu-Mn Alloys

- **Curie or Neel (T_c) Temperatures** when magnetic impurity concentration higher than 1%, experimental results showed the presence of Curie or Neel temperature (T_c) In the area between the maximum of resistivity (T_{max}) and the spin freezing temperature (T_f), which is evidence of the existence of direct exchange interaction between magnetic impurities. This result is inconsistent with EA and SK theories where the system directly turns of spin glass (SG) into paramagnetic (PM) phase, but theory of Wolfarth [52,53] or the so-called theory of clusters has explained the model of Neel [54,55] through free clusters, but without any interactions between those clusters, which will give a status similar to the paramagnetic, but by the clusters system and not by a singular spin especially in the case of low concentration, This phenomenon called super paramagnetic (SPM). It is assumed that the interactions among cluster are direct interactions (d-d),wherein the iron alloys give ferromagnetic (FM) regime and in the manganese alloys are antiferromagnetic (AFM). The decreasing temperature level will increase the interactions d-d to start a semi-ferromagnetic phase shows the temperature of Curie or Neel. This is the reason, as it's believed, why, experimentally, this temperature appears.

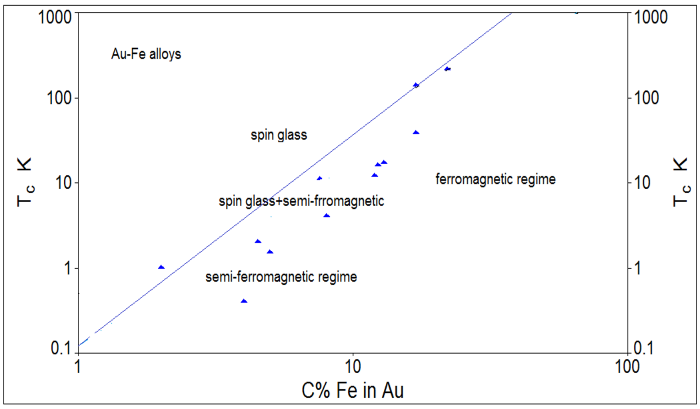

Figure 10: Critical Curie Temperatures as a Function of Concentrations Au-Mn Alloys

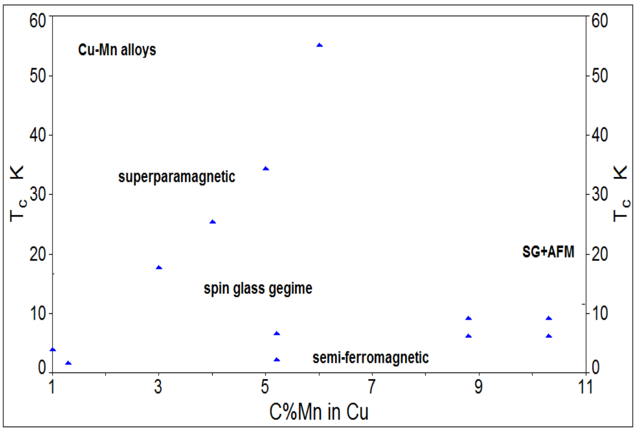

Figure 11: Critical Curie Temperatures as a Function of Concentrations Cu-Mn Alloys

- **Minimum Temperatures (T_{min})** there are two kinds of minimum temperatures, the first appears in Kondo alloys where these temperatures separate between Kondo regime and phonons regions at higher temperatures (T_{phonon}>T_K) [56]. The second appears in spin glass regime at temperatures more lower than the spin freezing temperature, a phase transition occurs through the emergence of spin waves which detected by experimental studies of resistivity and magnetic specific heat. Theory of (N. Rivier, and K. Adkins) RA [57, 58] has supported this attitude where the system enter into a semi- ferromagnetic mixed with spin glass which emphasizes the

appearance of Curie and Neel temperature (T_c) at that range of temperature. From Figure (12, 13) it is noted that these temperatures are positive (Curie temperature). It is evidence that the system trying to be in the phase of ferromagnetism.

Great accuracy studies [59] show that the spin freezing temperature, which discovered from the study of magnetic susceptibility, can appear above and below this temperature approved different relaxation times, which shows that this temperature is not a temperature of phase transition, but the situation is a sudden turning of the relaxation times to become very long (the memories) [60,61].

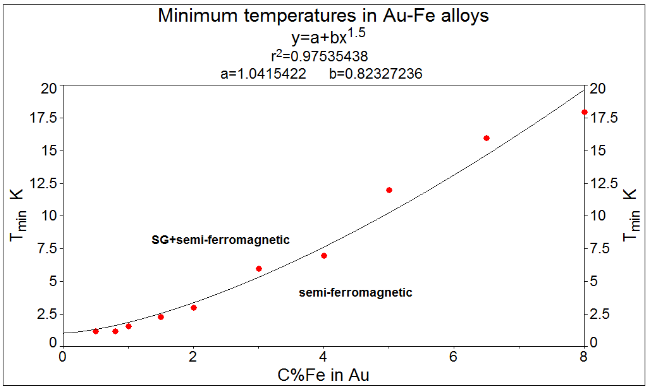

Figure 12: Minimum Temperatures as a Function of Concentrations in Au-Fe Alloys

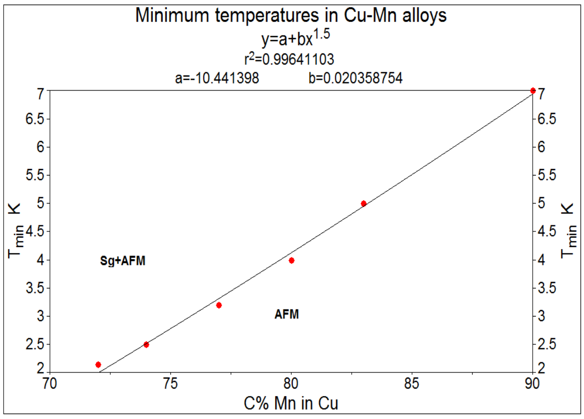

Figure 13: Minimum Temperatures as a Function of Concentrations in Cu-Mn Alloys

- At very low temperature, under half Kelvin, the interactions of hyperfine RKKY interactions begin, and the nucleus participates in a new type of spin glass (nuclear spin glass).

CONCLUSIONS

Theoretically, it's difficult to calculate exchange integral constant with easy methods, whereas Critical temperature - concentration diagrams give an excellent correlation with theoretical exchange integral constant in magnetic alloys, and the relation between critical temperatures as a function of concentrations identical with exchange integral constant, and need to calculate J_{xx} with high accuracy according to each regime.

The following scene of the critical points and their relationship with an exchange constant can be imagined as follows:

- T_k is related to J_{sd} where Kondo regime prevails.
- T_f is related to J_{RKKY} in spin glass Phase (SG) where RKKY interactions between clusters, or RKKY interactions between singlespins prevails.
- T_{max} is related to Paramagnetic phase (PM) and super paramagnetic phase (SPM) where Curie law prevails.
- T_c is related to J_e Mixed phase for concentration over 10% (Mictomagnetic), And with J_e Ferro or antiferromagnetic phase for concentration over 20% (FM).
- T_{min} is related to J_e, where semi-ferromagnetic phase (FM) begins to be formed, which thermally followed the spin glassregime (SG).
- T_{nf} is related to J_{hf}^{RKKY} where hyperfine indirect interactions appears below half Kelvin (Hyperfine-RKKY-interactions)

Complicated calculations are necessary to connect between theoretical exchange integral constant J_{xx} and critical points which will be expected to achieve in another paper soon.

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